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Understanding of transmission in the range of high-order photonic bands in thin opal film

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Diffraction in the face centered cubic lattice cannot explain some minima observed in the transmission spectra of self-assembled opal films. Here, we compared them with minima observed in the transmission spectra of a hexagonal close packed monolayer of spheres of the same diameter. The identity of the sphere packing on the surface of the opal film and in the sphere monolayer was demonstrated by the light diffraction at the sample surfaces. It was shown that excitation of surface propagating modes in the opal film is responsible for the formation of additional minima in opal film transmission. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2920443]

Thin films of colloidal crystals prepared by self-assembly of colloidal particles[1] are nowadays the most widely studied templates leading to three-dimensional (3D) photonic crystals (PhCs). Understanding of transmission in the spectral range of high-order bands in opal-based PhCs is vitally important since in this spectral range the omnidirectional photonic bandgap (PBG),[2,3] the anomalous refraction,[4] and the slow optical modes[5] take place. Meanwhile, experiments with light propagation in this range show that the observed spectral features do not exactly coincide with the PBG structure calculated for infinitely large crystal.[6,7] Hence a simple experiment to clarify this discrepancy is highly desirable.

It has been reported that the optical transmission in a monolayer of spheres exhibits minima occurring in a wavelength range $\lambda \sim D$, where D is a sphere diameter.[8,9] These minima were associated with excitation of optical eigenmodes of 2D PhC, which propagate in plane of a monolayer. The aim of this work is to demonstrate that similar effect can be observed in thin opal films.

Two types of samples were prepared from silica spheres of $D=519$ nm hydrophobized by using 3-(trimethoxysilyl) propyl methacrylate. Opal films were crystallized from these spheres by controlled evaporation from an ethanol solution on a glass substrate to form thin films with the [111] growth axis of the face centered cubic (fcc) lattice along the substrate normal.[10] A monolayer (1 ML) of the same spheres was deposited on a glass substrate by using the Langmuir–Blodgett (LB) technique.[11–13] As previously shown, 1 ML of spheres crystallize in a hexagonal lattice.[13] The LB approach was used because it has a distinct advantage over self-assembly for the preparation of samples with exact number of monolayers.

Angle-resolved transmission spectra were acquired at different angles of incidence θ, from 0° to 26° with respect to the film normal (inset Fig. 1). The collimated light beam of 1 mm in diameter from a tungsten halogen lamp was delivered through a prism polarizer to study the polarization anisotropy. The transmitted light was collected from a solid angle of 2°. A quarter-wavelength plate was used in front of the entrance slit of the spectrometer to transform the linear polarized signal into circularly polarized light. First order diffraction spectra were collected with the same beam setting as used for transmission measurements [inset Fig. 2(a)]. The incident angle was kept constant $\theta=50°$ and the angle of diffraction α was varied.

At $\theta=0°$, the transmission spectrum of the opal film shows a minimum centered at $\lambda_{111}=1019$ nm, a rapid drop of transmission toward a second minimum at 539 nm, and further structure at shorter wavelengths (Fig. 1). Remarkably, the opal film exhibits the minimum the $\lambda_{2D}=539$ nm, which occurs in the 1 ML sample. This observation leads us to

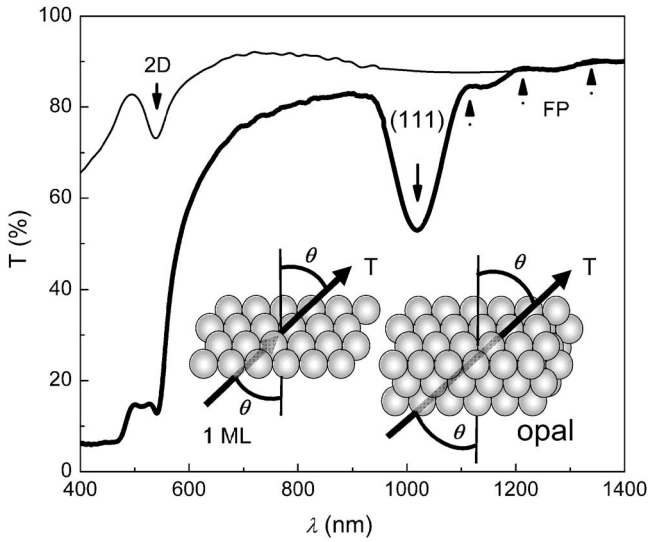


FIG. 1. Transmission spectra in *s*-polarized light of the 1 ML sample (thin line) and the opal film (thick line) assembled from 505 nm spheres at $\theta=0°$. The arrows indicate the transmission minima in the 1 ML sample (2D) and at the stack of (111) planes in the opal as well as the Fabry–Pérot oscillations of transmission in the opal film. The inset shows the measurement layout.

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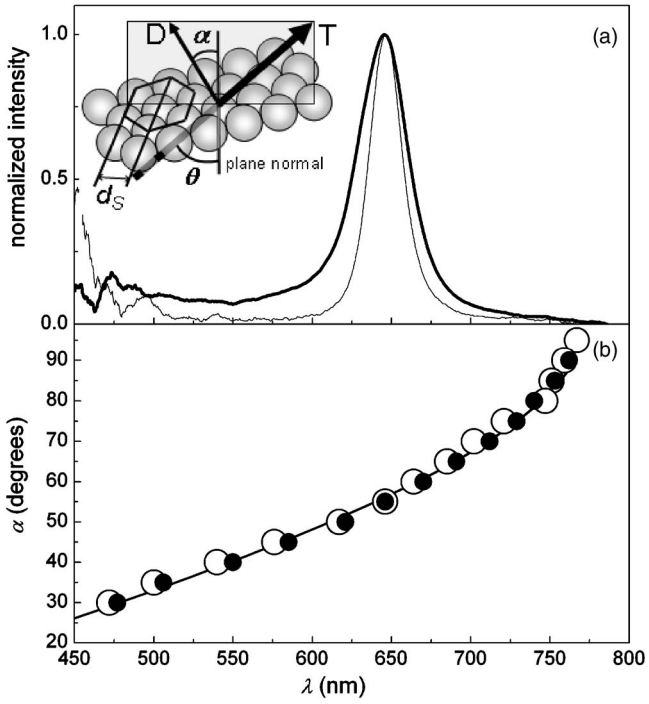


FIG. 2. (a) Surface diffraction spectra of the 1 ML sample (thin line) and the opal film (thick line). The inset shows the layout of the measurements: the plane of the light incidence and the corresponding lattice period. (b) Dispersion of the surface diffraction resonance of the 1 ML sample (empty circles) and in the opal film (solid circles) as a function of the angle of diffraction for $\theta=50°$. The lines show the fit to the surface diffraction expression. The inset explains the parameter d_S.

associate this feature with 2D eigenmodes in the 1 ML sample.

The lattice of the 1 ML LB film is the same as that of the surface of the opal film assuming that both represent 2D diffraction gratings.[14] The diffraction resonance obtained along the same directions of the incident and diffracted light appears at the same wavelength λ_S for both samples [Fig. 2(a)]. The higher relative bandwidth of the resonance in the opal film compared to a 1 ML sample, namely, 0.059 vs 0.036, can be associated with the misalignment of crystallites in the opal film. The angular dispersion of such resonances in both samples was fitted to the grating diffraction expression $\lambda_S=d_S[\sin(50°)+\sin(A+\alpha)]$, where d_S is the half-period of the trigonal lattice for the wave vector of incident light propagating along the ΓM direction in a Brillouin zone of 2D hexagonal lattice and $A\approx 11.2$ is a free constant. The fact that the same fit approximates very well both sets of data allows us to conclude that examined structures are similar. For the triangular lattice, the extracted distance is $d_S=437$ nm, which gives a sphere diameter $D=505$ nm. The advantage of such an approach is that the refractive index (RI) does not influence the result.

Transmission spectra of both samples obtained in the angle θ range from 0° to 26° are shown in Fig. 3. As the angle increases, the minimum in spectra of the 1 ML sample splits in two branches. The transmission surface of the opal film is also composed of two branches. In addition, the opal demonstrates a fine structure of minima and the presence of other branches. Similarity of angle dispersion of branches in the opal film and the 1 ML sample is further evidence of 2D mode contribution to absorption in the opal film.

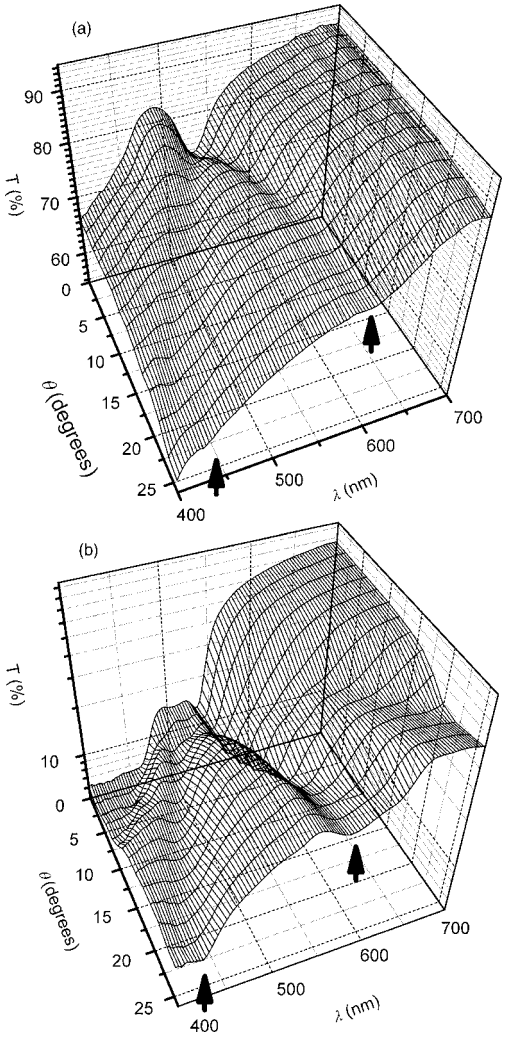


FIG. 3. The transmission spectra of the 1 ML sample (a) and the opal film (b) in the $\lambda\sim D$ spectral range. Arrows show the transmission minima with similar dispersions.

Detailed inspection of the dispersion of transmission minima in both samples is given in Fig. 4 for two orthogonal light polarizations. It is commonly accepted that transmission minima in the opal relate to the diffraction resonances at the crystal planes except the regions of the multiple diffraction.[15] Hence, the dispersion can be examined against the Bragg law $\lambda_{0\ hkl}=2d_{hkl}n_{\text{eff}}\sqrt{1-\sin^2 r_{hkl}}$, where $d_{hkl}=\sqrt{2}D/\sqrt{h^2+k^2+l^2}$ is the interplane distance along the $[hkl]$ direction, n_{eff} is the effective RI, and r_{hkl} is the internal angle between the incident beam and the $[hkl]$ direction. The external angle of incidence θ is related to the internal angle r_{111} via Snell's law: $n_{\text{eff}}\sin(r_{111})=n_{\text{air}}\sin(\theta)$. From fitting the angle dispersion of the (111) transmission minimum (Fig. 1) to the Bragg–Snell law using the sphere diameter obtained from the surface diffraction fit, the $n_{\text{eff}}=1.233$ is derived. The thickness of the opal film was estimated from the period of Fabry–Pérot oscillations of the transmission as ten (111) planes.

These estimates allow the dispersions of diffraction resonances for all relevant planes of the fcc lattice to be plotted (lines in Fig. 4). The dispersions of minima in the 1 ML sample provide another reference.

There are three strong minima observed in the opal spectra in *s*-polarized light represented by the o1, o3, and o4

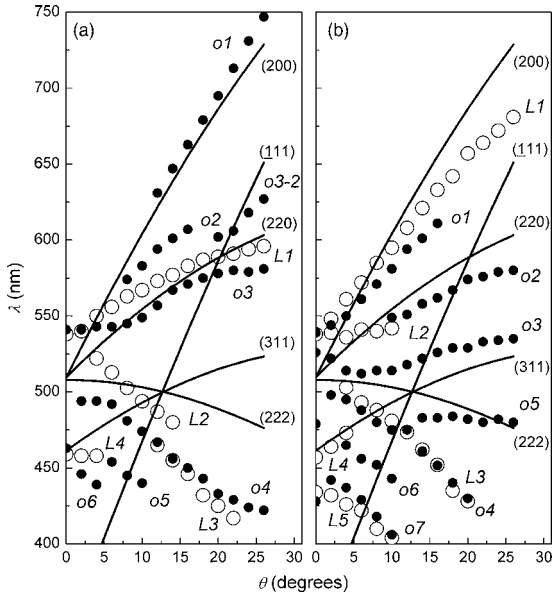


FIG. 4. The dispersion of transmission minima in the 1 ML sample (empty circles) denoted as L1, L2, … and the opal film (solid circles) denoted as o1, o2, … in the $\lambda \sim D$ spectral range. The lines show the dispersion of diffraction resonances in the fcc lattice and labeled with (*hkl*) indices of corresponding planes. (a) and (b) panels are for *s*- and *p*-polarized light.

branches [Fig. 4(a)]. The o1 branch unambiguously relates to the (200) diffraction in the fcc lattice. The o4 branch can be assigned to the L3 diffraction branch of the 1 ML sample. The origin of the o3 branch is not obvious. On the one hand, the o3 diffraction at low angles of incidence directly correlates to the L1 branch of in-plane resonances. On the other hand, at higher angles, the o3 resonances follow both the (220) diffraction in the fcc lattice and the L1 diffraction. The split of the o3 branch in o3 and o3-o2 branches at $\theta \geqslant 20°$ may be explained by the multiple diffraction simultaneously occurring at $(\bar{1}11)$ and (220) sets of planes in the fcc lattice. The o2, o5, and o6 branches refer to the shoulders in the transmission spectra. They also occur in the vicinity of 2D resonances but cannot be associated with any diffraction resonances in the fcc lattice. In the p-polarized light, the deep minima of the o1 branch follow the dispersion of the L1 minima in the 1 ML sample and degrade with the angle increase. The o2 and o3 branches follow the (220) and (311) resonances in the fcc lattice, respectively. The o4 branch is of mixed origin. At low angles, the associated minima are of moderate attenuation and follow the L3 dispersion, but at $\theta > 14°$, the o5 branch with stronger attenuation splits off and follows the (222) diffraction branch of the fcc lattice. Moreover, o3 and o5 dispersion branches reveal the avoided crossing. The o6 and o7 branches show the dispersions of shoulders in the transmission spectra.

The deviation of experimental diffraction dispersions from the Bragg law prescriptions obtained for n_{eff}=const can be related to (i) the deformation of the opal lattice due to the directional film growth,[16] (ii) the increase of the RI of silica by ~0.5% in a studied wavelength range, and (iii) the divergence of the group index in the spectral range of the directional PBG. Whereas the group index variation approaches high values,[17] it can hardly produce a noticeable change in the effective value of the RI after averaging over the whole solid angle. The latter statement corroborates the smooth appearance of the spectrum of the density of photon states obtained for the bare opal in the reduced frequency range $\sqrt{2}D/\lambda > 0.7$.[18]

It is worth noting that transmission minima associated with the excitation of surface modes occur in the opal independent of the light polarization. For example, the o2 branch in the *s*-polarized light and the o1 branch in the *p*-polarized light can be both associated with the L1 branch in the *p*-polarized light. Similarly, the o6 branch in the *s*-polarized light follows the dispersion of the L5 branch in the *p*-polarized light in the same manner as the o7 branch in the *p*-polarized light. This observation indicates that surface modes in the opal film are related to but not identical to the modes of the monolayer of spheres.

Similar reflectance bands were earlier experimentally observed and reproduced numerically,[19] but the difference between surface and volume modes was ignored in this analsis.

In summary, we have demonstrated that surface optical modes should not be neglected when considering the optical properties of PhCs based on thin opal films. The excitation of the evanescent surface modes leads to the essential attenuation of the light in the spectral range $\lambda \sim D$. The structure of these modes is similar to the PBG structure of the 2D PhC composed of single monolayer of spheres. The applied methodology, namely, the combination of surface and volume diffraction measurements, allows the independent determination of the lattice constant and the effective index of refraction of the 3D PhC.

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